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*Published in:*
EPRINTS-BOOK-TITLE

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*Document Version*
Publisher's PDF, also known as Version of record

*Publication date:*
1999

Link to publication in University of Groningen/UMCG research database

*Citation for published version (APA):*
Giessen, E. V. D., & Lai, J. (1999). Temperature Effects on Crack Tip Plasticity in Polymers. In *EPRINTS-BOOK-TITLE* University of Groningen, The Zernike Institute for Advanced Materials.

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# TEMPERATURE EFFECTS ON CRACK TIP PLASTICITY IN POLYMERS

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Abstract. The crack tip fields in amorphous glassy polymers are distinctly different from that in metals, and are associated with the initiation and propagation of shear bands. To study the effect of temperature on the near-tip behaviour for a stationary mode I crack, computations at low loading rates are confronted with adiabatic computations at high loading rates. It is shown that self-heating can raise the temperature in SAN sufficiently high locally to reach the glass transition temperature.

## 1. Introduction

Polymers, much like metals, exhibit a marked ductile-to-brittle transition when the loading rate is increased. There is growing experimental evidence, however, that for a number of polymeric materials there is an inverse transition at sufficiently high loading rates where the fracture toughness increases again. In particular, this is observed in amorphous polymers, such as polystyrene (PS) and PMMA which are commonly considered to be brittle (e.g. Fuller *et al.*, 1975), as well as in blends based upon amorphous matrix polymers (e.g. Steenbrink, 1997). This remarkable toughening effect is often referred to as thermal blunting and is generally attributed to adiabatic heating of the material caused by plastic deformation near the propagating crack. Direct and indirect observations have indicated that the temperatures can become high enough to cause local 'melting' of the material.

These intriguing experimental findings are supported qualitatively by estimates based on simple fracture mechanics considerations (e.g. Williams, 1972). Within an ongoing research programme on the fracture of polymers and polymer blends, we are addressing the same issue in more detail at a somewhat smaller length scale. One of the driving forces is the finding that



*O. T. Bruhns and E. Stein (eds.),*
*IUTAM Symposium on Micro- and Macrostructural Aspects ofThermoptasticity,* 167–176.
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the plastic zones in amorphous glassy polymers are distinctly different from the well-known kidney-shaped plastic zones that are usually presumed in fracture mechanics and copied from the plastic zones in metals. As demonstrated previously (Lai and Van der Giessen, 1997), this difference draws from the essential difference between the elastic-plastic response of polymers and that of metals. Upon yield, metals tend to exhibit a gradually decreasing strain hardening, whereas glassy polymers show the opposite behaviour of intrinsic softening upon yield, followed by progressive hardening. As a direct consequence, crack tip plasticity in polymers is caused by the initiation and propagation of shear bands.

The objective of the present paper is to supplement the previous isothermal studies of the crack tip fields around a stationary mode I crack in glassy polymers with computations that assume the opposite limit of local adiabatic conditions, and which account for the temperature dependence of the viscoplastic behaviour of these materials. In particular we are interested to see how the temperature rise develops while the plastic zones propagate in the form of shear bands. The glass transition temperature is set as the upper limit in the simulations, since the material becomes rubbery above this temperature. Crack growth by crazing is not considered here.

## 2. Material Model

### 2.1. BASIC CONSTITUTIVE MODEL

The deformation mechanisms that we account for in the present considerations are: (i) thermoelasticity, (ii) temperature and pressure dependent yield, (iii) intrinsic strain softening, and (iv) progressive strain hardening. Plastic flow in amorphous polymers is caused by collective segmental rotations taking place in the molecules, and it is the collectivity that is responsible for the intrinsic softening immediate upon yield. The strain hardening is caused by the subsequent stretching of the network formed by the entangled molecules (Haward, 1973).

It follows that there are two contributions to the free energy of a material element. Firstly, there is the free energy associated with thermoelastic processes, which are of intramolecular nature (atomic interactions within molecules), and for which the free energy function $f^{\mathrm{e}}$ (per unit mass) is a function of temperature $T$ and some appropriate elastic strain tensor. The second contribution is associated with the stretching of molecular chains between the entanglements, which causes the progressive strain hardening during plastic flow. This free energy contribution is primarily of an entropic nature (similar to that of a rubber-elastic network; see Treloar, 1975). The corresponding free energy function, $f^{\mathrm{p}}$, is taken to be a function of $T$ and of plastic strain. Since this energy contribution is of a completely different

physical nature as the thermoelastic free energy, we assume that they are uncoupled, so that the total specific free energy is given as $f = f^{e} + f^{p}$.

The constitutive theory can now be formulated consistently within the standard framework of continuum thermomechanics, but space limitations force us to focus on the final constitutive equations. First of all, we obtain thermodynamic state equations for entropy $\eta$, the Cauchy stress $\boldsymbol{\sigma}$ and the thermodynamic stress $\boldsymbol{b}$ associated with the plastic strain dependence of $f^{p}$. When we assume small elastic strains and small changes of the temperature relative to a reference temperature $T_0$, the equations of state for $\boldsymbol{\sigma}$ can be differentiated with respect to the time, and after appropriate manipulations can be rewritten in the rate form

$$\overset{\nabla}{\boldsymbol{\sigma}} = \boldsymbol{\mathcal{L}}^{e}\boldsymbol{D}^{e} - C\alpha_{c}\dot{T}\boldsymbol{I}\,. \tag{1}$$

Here, $\boldsymbol{\mathcal{L}}^{e}$ is the standard fourth-order isotropic elastic modulus tensor in terms of Young's modulus $E$ and Poisson's ratio $\nu$, $C = E/3(1-2\nu)$ is the bulk modulus ($G = E/2(1+\nu)$ is the elastic shear modulus) and $\alpha_{c}$ is the coefficient of cubic thermal expansion. Furthermore, $\overset{\nabla}{\boldsymbol{\sigma}} = \dot{\boldsymbol{\sigma}} - \boldsymbol{W}\boldsymbol{\sigma} + \boldsymbol{\sigma}\boldsymbol{W}$ is the Jaumann derivative of Cauchy stress based on the continuum spin tensor $\boldsymbol{W}$, and $\boldsymbol{D}^{e}$ is the elastic part of the stretching tensor,

$$\boldsymbol{D} = \boldsymbol{D}^{e} + \boldsymbol{D}^{p}\,. \tag{2}$$

The stress tensor $\boldsymbol{b}$ is determined by the plastic strain dependence of $f^{p}$ through $\rho\dot{f}^{p} = \boldsymbol{b}\cdot\boldsymbol{D}^{p} + T\partial f^{p}/\partial T$.

It has been shown by Wu and Van der Giessen (1993) that the plastic free energy $f^{p}$ of the entangled network can be obtained from the free energy expression of individual non-Gaussian molecular chains (see Treloar, 1975) by an appropriate averaging procedure. It has also been demonstrated that the numerical results of such averaging can be accurately approximated by a nonlinear combination of the stresses according to two simplified network models:

$$\boldsymbol{b} = (1-\rho)\boldsymbol{b}^{3\text{-ch}} + \rho\boldsymbol{b}^{8\text{-ch}}\,. \tag{3}$$

The constitutive equations for the three-chain and eight-chain stresses $\boldsymbol{b}^{3\text{-ch}}$ and $\boldsymbol{b}^{8\text{-ch}}$ contain only two material parameters: the initial shear modulus $C^{R} \equiv nkT$ ($n$ is the volumetric density of entanglements, $k$ is Boltzmann's constant) and the number of segments per chain, $N$, which implies a limit stretch of $\sqrt{N}$. The weight factor $\rho$ in (3) is $\rho = 0.85\bar{\lambda}/\sqrt{N}$ with $\bar{\lambda}$ the maximum principal plastic stretch.

Substitution of the above free energy functions into the Clausius–Duhem inequality yields the mechanical dissipation $D$ as

$$\mathcal{D} = \bar{\boldsymbol{\sigma}}'\cdot\boldsymbol{D}^{p}\,, \quad \bar{\boldsymbol{\sigma}} = \boldsymbol{\sigma} - \boldsymbol{b}\,, \tag{4}$$

which shows that $\boldsymbol{b}$ serves as a back stress (the $'$ denotes the deviatoric part). This suggests to let the plastic strain rate tensor $\boldsymbol{D}^{\mathrm{p}}$ be defined through a dissipation function that is primarily dependent on the driving stress $\bar{\boldsymbol{\sigma}}'$. Assuming that yield in an amorphous polymer is an isotropic process and invoking normality, we find that

$$\boldsymbol{D}^{\mathrm{p}} = \frac{\dot{\gamma}^{\mathrm{p}}}{\sqrt{2}\,\tau}\bar{\boldsymbol{\sigma}}', \quad \tau = \sqrt{\tfrac{1}{2}\bar{\boldsymbol{\sigma}}' \cdot \bar{\boldsymbol{\sigma}}'}, \quad \dot{\gamma}^{\mathrm{p}} = \sqrt{\boldsymbol{D}^{\mathrm{p}} \cdot \boldsymbol{D}^{\mathrm{p}}}, \tag{5}$$

with $\dot{\gamma}^{\mathrm{p}} = \dot{\gamma}^{\mathrm{p}}(\tau, T; p)$. The particular form we use in this work is based on Argon's (1973) model for the temperature and rate dependent yield of amorphous polymers, in which

$$\dot{\gamma}^{\mathrm{p}} = \dot{\gamma}_0 \exp\left[-\frac{A s_{\mathrm{y}}}{T}\left(1 - \left(\frac{\tau}{s_{\mathrm{y}}}\right)^{5/6}\right)\right]. \tag{6}$$

and $s_{\mathrm{y}} = s + \alpha p$ ($\dot{\gamma}_0$, $A$ and $\alpha$ are material parameters). Here, $s$ is the athermal shear strength which is taken to evolve with strain according to

$$\dot{s} = h(1 - s/s_{\mathrm{ss}})\dot{\gamma}^{\mathrm{p}}, \tag{7}$$

as a phenomenological description of strain softening upon yield ($h$ is the initial softening modulus and $s_{\mathrm{ss}}$ is the ultimate minimal shear resistance).

Under adiabatic conditions, the entropy balance yields $T\rho\dot{\eta} = \mathcal{D}$. From (4) and from the state equation $\eta = -\partial f/\partial T$ we then infer that

$$-\rho T \frac{\partial^2 f}{\partial T^2}\dot{T} = \bar{\boldsymbol{\sigma}}' \cdot \boldsymbol{D}^{\mathrm{p}} \quad \text{with } \frac{\partial^2 f^{\mathrm{e}}}{\partial T^2} = -\frac{c_{\mathrm{v}}}{T}, \quad \frac{\partial^2 f^{\mathrm{p}}}{\partial T^2} = 0 \tag{8}$$

for the free energy functions referred to above, assuming small temperature changes and neglecting latent heat contributions.

## 2.2. THERMAL EFFECTS

Polymers are intrinsically strongly dependent on temperature, even in the glassy range $T < T_{\mathrm{g}}$. The thermoelastic properties of glassy polymers are among the least temperature sensitive properties, except when the temperature is closely approaching $T_{\mathrm{g}}$. As the above constitutive equations are appropriate only up to $T_{\mathrm{g}}$ anyway, we here neglect the temperature dependence of the thermoelastic properties.

The viscoplastic behaviour of amorphous plastics depends on temperature in a number of ways. First of all, experiments show that the yield stress falls almost linearly with temperature up to temperatures quite close to the glass transition temperature $T_{\mathrm{g}}$ (e.g., Bowden, 1973; Haward, 1973). This

effect is already incorporated in the Argon yield model in (6). Ignoring for simplicity the factor 6/5 in the stress dependence, one immediately finds that $d\tau/dT \approx -1/A \ln(\dot{\gamma}_0/\dot{\gamma}^{\mathrm{p}})$, showing that the temperature dependence at a constant strain rate $\dot{\gamma}^{\mathrm{p}}$ is governed solely by $A$. Softening, in general, is also dependent on temperature. However, some materials, including the one used in the calculations to be presented later, do not exhibit this (Steenbrink, 1997).

Experiments show that the intensity of hardening decreases with increasing temperature while the finite stretchability increases. Raha and Bowden (1972) have suggested that this is caused by thermal dissociation of the entanglements that form the molecular network. Based on this idea, they suggest that the network density $n$ (defining the hardening modulus $C^{\mathrm{R}}$) depends on temperature according to

$$n(T) = B - D \exp\left(-E_{\mathrm{a}}/RT\right) , \tag{9}$$

while $nN$ = const because the numbers of molecular links remains constant. In(9), $E_{\mathrm{a}}$ is the dissociation energy, $R$ is the gas constant while $B$ and $D$ are material parameters. By imposing the reasonable side condition that the network 'breaks down' when the glass transition is approached, $n(T_{\mathrm{g}}) = 0$, we find that these parameters need to satisfy $B/D = \exp\left(-E_{\mathrm{a}}/RT_{\mathrm{g}}\right)$ .

## 3. Small-scale yielding problem

Following the isothermal study of Lai and Van der Giessen (1997), we investigate the development of plasticity near the blunted crack tip in a glassy polymer under mode I loading. We confine attention to small-scale yielding, and use the boundary layer approach for a stationary, plane strain crack. Remote from the crack tip, the elastic displacement rates are prescribed in accordance with the chosen rate of change of the applied stress intensity factor $K_{\mathrm{I}}$. Either isothermal conditions are assumed, or, when $\dot{K}_{\mathrm{I}}$ is much higher, adiabatic conditions in the tip region. In the latter case, also heat transport to the crack faces is neglected, so that these conditions imply a real upper limit to the temperature rise near the tip.

## 4. Results

We present results here for a particular material, namely styrene-acrylonitrile(SAN; $T_{\mathrm{g}} = 383\,\mathrm{K}$) which is the matrix base material for the well-known blend ABS. Uniaxial compression experiments on cylindrical specimens have been carried out by Steenbrink (1997) at a range of temperatures and strain-rates in order to obtain the elastic and yield properties as $E/s_0 = 12.5$, $\nu = 0.38$, $s_0 = 120\,\mathrm{MPa}$, $A = 129\,\mathrm{K/MPa}$, $\dot{\gamma}_0 = 1.06 \times 10^8\,\mathrm{s}^{-1}$,

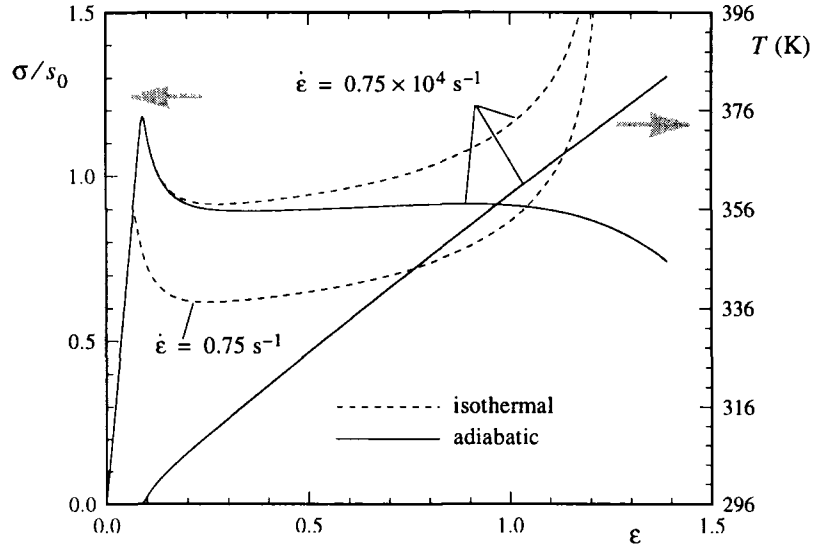


*Figure 1.* True stress vs. true strain response of SAN to uniaxial tension under isothermal or adiabatic conditions, including the corresponding temperature development.

$\alpha = 0.25$, $h/s_0 = 12.5$, $s_{ss}/s_0 = 0.79$. The Young's modulus $E$ is taken here to match the yield strain, so as to circumvent the viscoelastic effects prior to yield. The hardening parameters at room temperature ($T_0 = 296\,\mathrm{K}$) are found to be $C^R = 4\,\mathrm{MPa}$ and $N = 12$, while the dissociation energy is estimated as $E_a/R = 2.8 \times 10^3\,\mathrm{K}$. The cubic thermal expansion coefficient is taken to be the same as for PS, $\alpha_c = 2 \times 10^{-4}\,\mathrm{K}^{-1}$. Finally, we take $\rho = 1.08 \times 10^3\,\mathrm{kg/m^3}$ and $c_v = 1.38 \times 10^3\,\mathrm{J/kgK}$.

For these parameters, Fig. 1 shows the uniaxial true stress-strain response at two different strain-rates that differ by four orders of magnitude. If isothermal conditions are assumed, an increase of the strain-rate by a factor of four is essentially shifts the entire response to higher stress levels without changing the softening and hardening characteristics. When instead it is imagined that the largest strain-rate is high enough to cause adiabatic heating, both the effective yield stress and the network resistance drop with continued straining. In effect, the temperature is found to rise so high (see also Fig. 1) that re-hardening does not occur at all before the glass transition temperature ($T_g = 383\,\mathrm{K}$) is reached.

Figure 2 depicts the development of the instantaneous plastic zone around the crack tip under isothermal conditions. The plastic zone is visualized here in terms of the distribution of the current value of the plastic shear rate $\dot{\gamma}^p$. As expected from the parameter study carried out previously (Lai and Van der Giessen, 1997), the material corresponding to Fig. 1 exhibits a

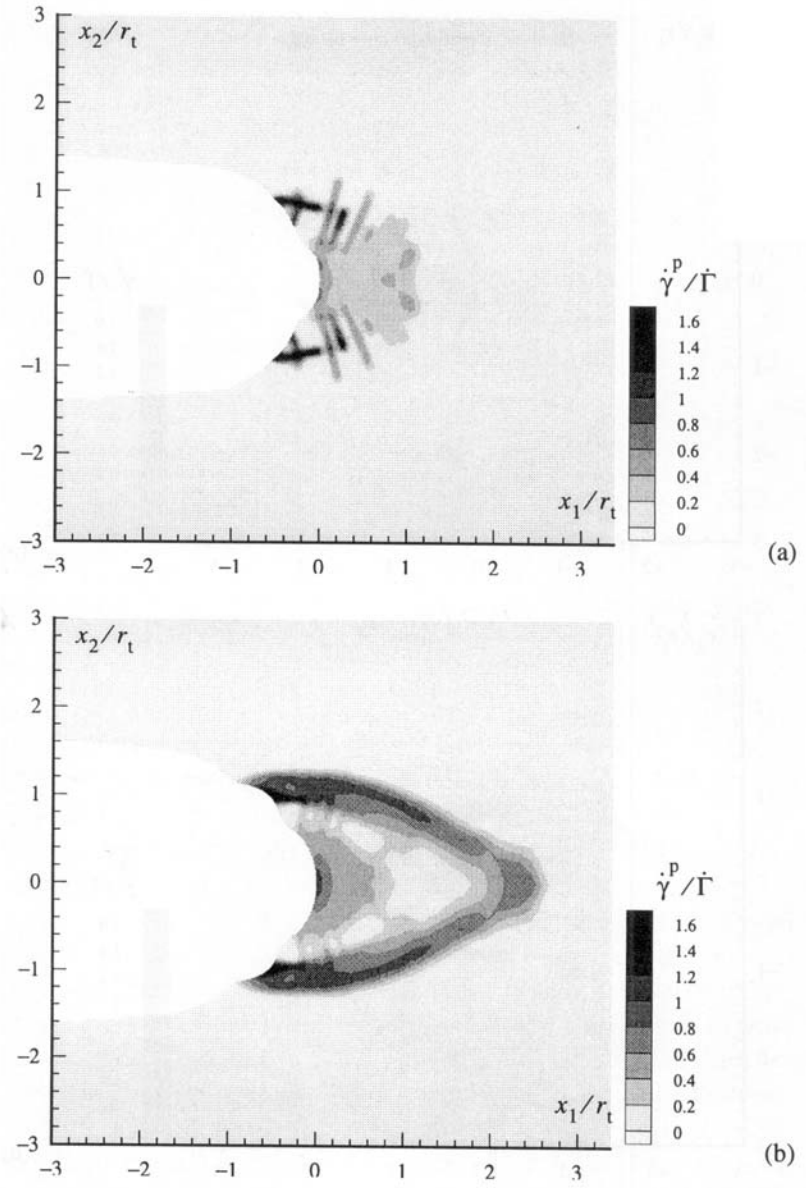


*Figure 2.* Distribution of instantaneous plastic shear rate $\dot{\gamma}^p$ near the crack tip at a loading rate of $\dot{K}_I = 1.1\ \mathrm{MPa}\sqrt{\mathrm{m}}/\mathrm{s}$ under isothermal conditions: (a) $K_I = 2.31\ \mathrm{MPa}\sqrt{\mathrm{m}}$; (b) $K_I = 3.38\ \mathrm{MPa}\sqrt{\mathrm{m}}$.

plastic zone that comprises a family of shear bands emanating from the tip surface at sufficiently low levels of the applied stress intensity (Fig. 2.a). At

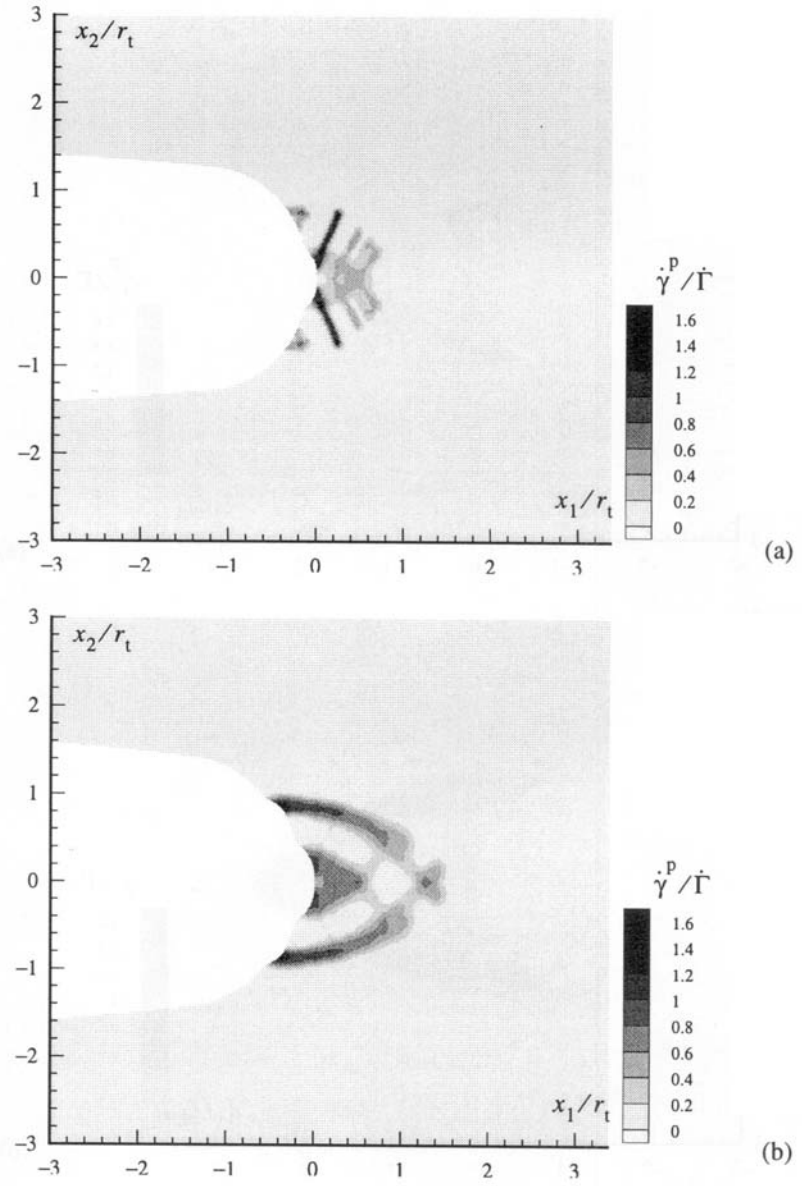


*Figure 3.* Distribution of instantaneous plastic shear rate $\dot{\gamma}^p$ near the crack tip at a loading rate of $\dot{K}_I = 1.1 \times 10^4\ \mathrm{MPa}\sqrt{\mathrm{m}}/\mathrm{s}$ under adiabatic conditions: (a) $K_I = 2.45\ \mathrm{MPa}\sqrt{\mathrm{m}}$; (b) $K_I = 3.34\ \mathrm{MPa}\sqrt{\mathrm{m}}$ (cf. Fig. 4).

increasing loading, continued propagation of the shear bands takes place with eventually two main shear bands (Fig. 2.b).

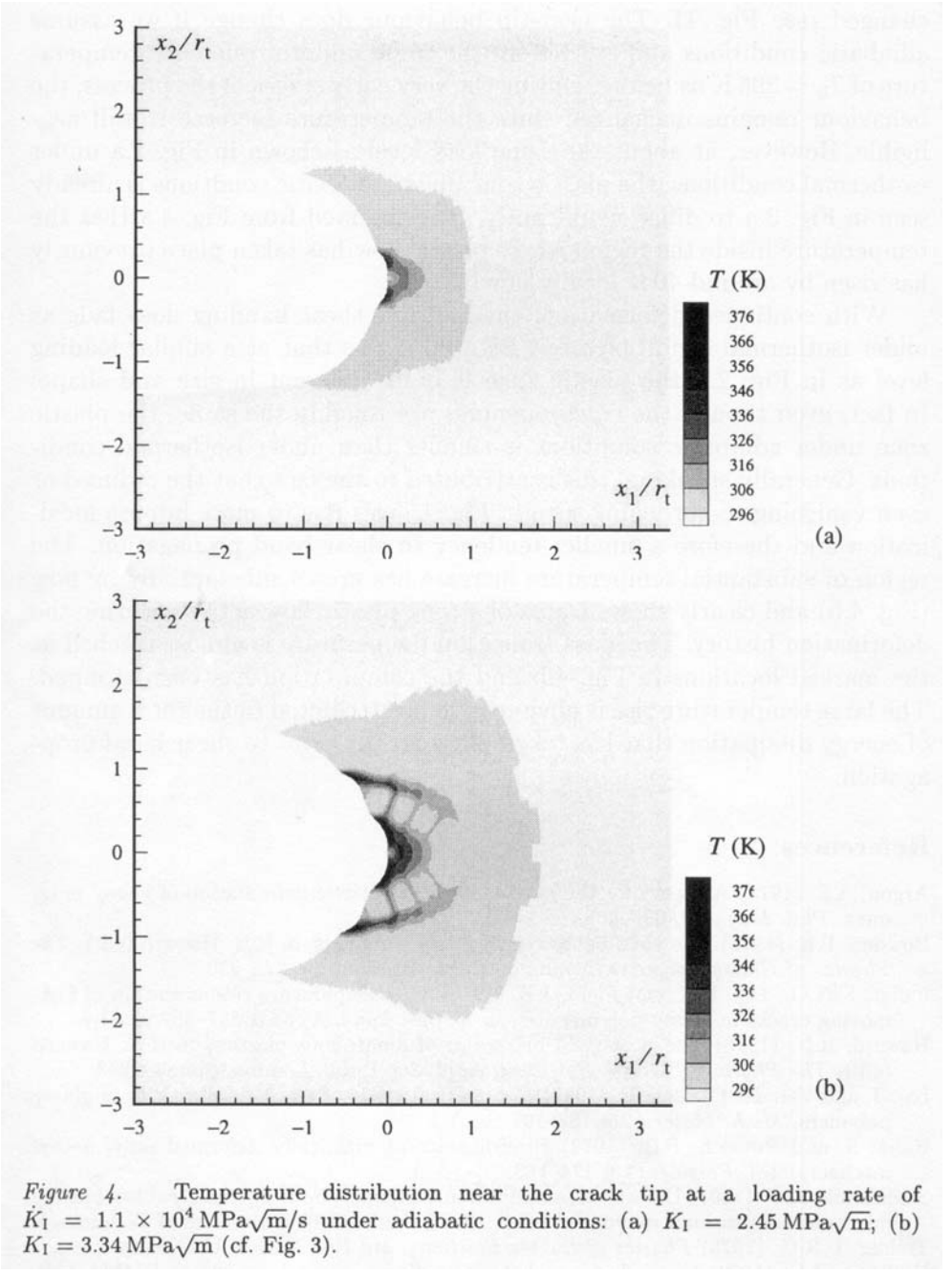


*Figure 4.* Temperature distribution near the crack tip at a loading rate of $\dot{K}_I = 1.1 \times 10^4\,\mathrm{MPa}\sqrt{\mathrm{m}}/\mathrm{s}$ under adiabatic conditions: (a) $K_I = 2.45\,\mathrm{MPa}\sqrt{\mathrm{m}}$; (b) $K_I = 3.34\,\mathrm{MPa}\sqrt{\mathrm{m}}$ (cf. Fig. 3).

At a four times higher loading rate, similar plastic zones will develop if the process is still isothermal, since the response is qualitatively un-

changed (see Fig. 1). The near-tip behaviour does change if we assume adiabatic conditions and start from the same uniform reference temperature of $T_0 = 296\,\mathrm{K}$ as before. During the very early stages of the process, the behaviour remains unchanged since the temperature increase is still negligible. However, at about the same load level as shown in Fig. 2.a under isothermal conditions, the plastic zone under adiabatic condtions is already seen in Fig. 3.a to differ significantly. It is observed from Fig. 4.a that the temperature inside the region where plastic flow has taken place previously has risen by around 40 K locally now.

With continued deformation, the multiple shear banding does fade as under isothermal conditions, but Fig. 3.b shows that at a similar loading level as in Fig. 2.b the plastic zone is quite different in size and shape. In fact, even though the crack openings are roughly the same, the plastic zone under adiabatic conditions is smaller than under isothermal conditions. Generally speaking, this is attributed to the fact that the reduced or even vanishing re-hardening seen in Fig. 1 gives rise to more intense localization and therefore a smaller tendency to shear band propagation. The region of substantial temperature increase has grown substantially by now (Fig. 4.b) and clearly shows traces of strong plastic flow activity during the deformation history. The glass transition temperature *is* almost reached at the marked locations in Fig. 4.b and the computation has been stopped. The large temperature rise is obviously to be attributed to the total amount of energy dissipation that has taken place locally prior to shear band propagation.

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